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Did policies to abate atmospheric emissions from traffic have a positive effect in London?☆

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ABSTRACT

A large number of policy initiatives are being taken at the European level, across the United Kingdom and in London to improve air quality and reduce population exposure to harmful pollutants from traffic emissions. Trends in roadside increments of nitrogen oxides (NO_X), nitrogen dioxide (NO_2), particulate matter (PM), black carbon (CBLK) and carbon dioxide (CO_2) were examined at 65 London monitoring sites for two periods of time: 2005–2009 and 2010–2014. Between 2005 and 2009 there was an overall increase in NO_2 reflecting the growing evidence of real world emissions from diesel vehicles. Conversely, NO_2 decreased by $10\% \cdot year^{-1}$ from 2010 onwards along with $PM_{2.5}$ ($-28\% \cdot year^{-1}$) and black carbon ($-11\% \cdot year^{-1}$). Downwards trends in air pollutants were not fully explained by changes in traffic counts therefore traffic exhaust emission abatement policies were proved to be successful in some locations. PM_{10} concentrations showed no significant overall change suggesting an increase in coarse particles which offset the decrease in tailpipe emissions; this was especially the case on roads in outer London where an increase in the number of Heavy Good Vehicles (HGVs) was seen. The majority of roads with increasing NO_X experienced an increase in buses and coaches. Changes in CO_2 from 2010 onwards did not match the downward predictions from reduced traffic flows and improved fleet efficiency. CO_2 increased along with increasing HGVs and buses. Polices to manage air pollution provided differential benefits across London's road network. To investigate this, *k*-means clustering technique was applied to group roads which behaved similarly in terms of trends to evaluate the effectiveness of policies to mitigate traffic emissions. This is the first time that London's roadside monitoring sites have been considered as a population rather than summarized as a mean behaviour only, allowing greater insight into the differential changes in air pollution abatement policies.

1. Introduction

The air pollution close to roads in large urban areas is usually affected by emissions from traffic such as nitrogen oxides (NO_X), particulate matter (PM) and black carbon (CBLK) among other pollutants (Sundvor et al., 2012). Tail-pipe emissions from traffic are dominated by diesel engines which emit NO_X in form of nitrogen monoxide (NO) and primary nitrogen dioxide (NO_2) (Carslaw et al., 2011) causing steep spatial gradients close to roads (Carslaw and Beevers, 2005). Diesel vehicles also emit elemental carbon (EC) and CBLK (Mansfield et al., 1991), fine and ultrafine particles (particles with <2.5 μm and <0.1 μm in diameter, respectively) which can be inhaled deeper into the lung and therefore are thought to be more toxic than larger particles (HEI, 2013). Moreover, other non-exhaust traffic-related emissions such as resuspension, tyre-wear and brake-wear could represent an important fraction of coarse PM on roads (particles with >2.5 μm) (Amato et al., 2016). Exposure to traffic-related pollutants can be very considerable alongside urban roads in central areas, along retail and popular streets used by pedestrians. Adverse health effects associated with proximity to roads have been observed due to higher concentrations of individual or combinations of traffic-related pollutants (WHO, 2013). The mortality burden of NO_2 and $PM_{2.5}$ in London in 2010 has been estimated as equivalent to 9416 deaths at typical ages (Walton et al., 2015). Recent studies have found positive associations between EC and CBLK from diesel exhaust emissions and respiratory mortality (Atkinson et al., 2015). Toxicological research increasingly indicates that non-exhaust pollutants could also be responsible for some of the observed

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health effects (WHO, 2013). Traffic sources also release gases including carbon dioxide (CO_2) that have an impact on climate (IPCC, 2013). Anthropogenic sources of CO_2 in urban areas are related to combustion processes such as burning of fossil fuels and electricity production. Roadside increments of CO_2 can be taken as indication of direct exhaust emissions from traffic (Gratani and Varone, 2014).

A suite of policies have been implemented to improve air quality and reduce population exposure. The Euro emission standards were introduced in the early 1990s to reduce exhaust emissions from new vehicles and tighter standards have been introduced in the last two decades. Transport for London (TfL) invested in a program to fit a catalytic diesel particulate filters (CDPF) to its older buses by the end of 2005 (Carslaw and Beevers, 2005). By 2014 TfL completed a second bus retrofit campaign with over 1000 Euro III buses fitted with a Selective Catalytic Reduction (SCR) system to reduce NO_X emissions. Fitting SCR was prioritized for those buses with routes along busy roads in central London (TfL, 2014a). Other initiatives across London include the Low Emission Zone implemented in 2008 which limited the entrance of the most polluting diesel Heavy Good Vehicles (HGVs) in London. The Mayor's Air Quality Strategy in 2010 planned the roll out of new hybrid buses and low-emission buses (Euro IV) (GLA, 2010). All these policies have been accompanied by many local-scale schemes implemented by the London's boroughs. The ensemble of these initiatives is expected to have a direct impact on the air quality in the whole of London but especially alongside busy roads. Whilst the vast majority of roadside locations in London met the PM_{10} EU Annual Mean Limit Value of 40 $\mu g\ m^{-3}$ in 2014, the majority still exceeded the NO_2 EU Limit Value of 40 $\mu g\ m^{-3}$ by a large margin (Mittal et al., 2016). Additionally, meeting the EU $PM_{2.5}$ exposure reduction target remains challenging.

Evaluating the success of policies in improving air quality can be done though the study of trends in atmospheric concentrations in time (e.g. Gualtieri et al., 2014). Most of the approaches calculate trends in concentrations using one monitoring site representative of a given exposure scenario (e.g. roadside site); or by averaging the concentrations from a group of similar sites. In large urban settings, such as London, trends in air quality monitoring sites have been classified by their distance to the city centre, for instance, inner and outer London (Beevers et al., 2012). Other studies considered the variability of trends observed across a network. Colette et al. (2015) calculated trends in air pollutants in Europe using the AIRBASE network and a probability density function to summarize the trend distribution. Other recent approaches include the estimation of trends in air pollutants from satellite-based observations. These benefit from their wide spatial coverage, allowing calculation of trends at the continental scale (van der A. et al., 2008), country-scale (e.g. Cuevas et al., 2014) or at various urban centres worldwide (Schneider et al., 2015). Although these methods are valid and useful, these approaches can mask a wide heterogeneity in the impact of policies across an urban area and especially in roadside locations where emissions have a large spatial and temporal variability.

Our study was designed to evaluate the success of the policies to reduce the air pollution concentrations in London with special focus on traffic emissions. We benefitted from the large number of monitoring stations in the urban agglomeration (more than 100 monitoring stations in 2014) and trends of air pollutants concentrations were calculated for all 65 roadside sites available between 2005 and 2014. Due to the spatial representativeness of air quality monitoring stations, the duration and diversity of data, the London air quality database offers an unprecedented and effective way to analyze trends in surface air pollutants concentrations. The overall trend of air pollutants in London was calculated using statistical approaches used in meta-analysis studies that consider individual and population-wide variability. Roadside locations were grouped according to recent changes in air pollution concentrations and trends were related to specific policies and to changes in traffic counts and composition. This approach would be applicable to other cities with a large network of monitoring sites, and also at the country, region and worldwide scale.

2. Materials and methods

2.1. Monitoring sites

Measurements of NO_X, NO_2, particulate matter with aerodynamic diameter <10 μm (PM_{10}) and <2.5 μm ($PM_{2.5}$), black carbon (CBLK) and carbon dioxide (CO_2) were extracted from the UK Automatic Urban and Rural Network (AURN) and the London Air Quality Network (LAQN). These comprised 65 roadside Air Quality Monitoring Sites (AQMSs) (Supplementary Fig. 1). Note that some sites had collocated PM instruments measuring by different methods, e.g. Marylebone Road where PM was monitored by both TEOM and TEOM-FDMS. To distinguish these apart, the methods were assigned to different site codes (i.e. MY1 and MY7, respectively). Three roadside AQMSs in the London network measured CO_2 and CBLK for the period 2010–2014.

Measurements from Kensington and Chelsea - North Kensington (KC, 51.521°N, −0.2135°E) were taken as background concentrations (Supplementary Fig. 1). KC was chosen as background for three reasons: i) the use of a single background site allowed roadside increments to be directly compared between different roadside locations; ii) it is the urban background AQMS with the longest complete time series for all pollutants; iii) trends observed at KC were the same (within 2σ confidence interval) of the overall trends observed for all urban background sites in London (Supplementary Figs. 2 and 3), with the only exception being trends for NO_2 between 2005 and 2009 when a faster decrease (−1.07 $\mu g\ m^{-3}\ year^{-1}$) was observed compared with the other urban background sites (−0.37 $\mu g\ m^{-3}\ year^{-1}$).

The distance to London's city centre was calculated for each AQMS, setting the centre at Charing Cross (51.508°N, 0.125°W). Sites <10 km from Charing Cross were considered inner London; sites >10 km away from Charing Cross were considered outer London.

2.2. Measurements

NO_X ($NO + NO_2$) was measured by chemi-luminiscence and fortnightly calibrations enabled the traceability of measurements to national metrological standards. PM_{10} and $PM_{2.5}$ were measured by TEOM-FDMS (Tapered Element Oscillating Microbalance - Filter Dynamics Measurement System); by TEOM and by MetOne BAM (Beta Attenuation Monitors). TEOM-FDMS measurements were considered equivalent to the EU reference method, which is based on 24-h sampling and gravimetric analysis. PM_{10} measurements made by TEOM were converted to reference equivalent using the Volatile Correction Model (VCM) (Green et al., 2009). $PM_{2.5}$ measurements by TEOM were not corrected to reference equivalent as there is currently no agreed method for this. PM measurements by BAM were corrected to EU Reference equivalent using a factor of 1/1.2 (DEFRA, 2010). CBLK in $PM_{2.5}$ was measured by the Magee Aethalometer AE22 and raw data was corrected for the filter loading effect (Virkkula et al., 2007; Butterfiled et al., 2013). All instruments were subject to twice yearly audit tests by the National Physical Laboratory or Ricardo AEA.

CO_2 concentrations were measured using a LiCOR-820 Non-Dispersive IR analyzer. Two-point calibrations were carried out every 15 days with a zero-scrubber (soda lime) and a CO_2 standard gas referenced to the International Scale (WMO-X2007).

2.3. Traffic data

Street-level Annual Average Daily Flow (AADF) was obtained from the Department for Transport (DfT). AADF accounts for the annual average number of vehicles passing a point in the road network each day (vehicles day^{-1}) and it was available for different vehicle categories: cars and taxis, motorcycle, buses and coaches, light good vehicles (LGVs) and heavy goods vehicles (HGVs). Traffic data was obtained for the nearest traffic count point to each AQMSs that measured both NO_X and PM_{10} (Supplementary Fig. 1).

2.4. Trend analysis

This study used new statistical approaches to analyze the trends in air pollutants in a megacity. These were based in the concept of population commonly used in epidemiological and ecological studies. Here we used a population of monitoring sites in order to examine the temporal trends of air pollutants in London, taking into account both individual and population-wide variability.

To focus on the changes in ambient air pollution due to local traffic emissions (exhaust and non-exhaust), trends in roadside increments above the urban background concentration (denoted with Δ) were calculated. In this way changes over time due to processes at the regional scale (such a meteorological conditions, boundary layer dynamics, policies outside the city, etc.) did not confound the analysis.

Trends in roadside increments were calculated for two periods: 1st January 2005 to 31st December 2009; and 1st January 2010 to 31st December 2014. Trends between 2010 and 2014 were compared with those between 2005 and 2009 to discuss possible changes due to specific policies. The 2005 starting year was chosen as the first year for which TEOM-VCM measurements were available. Also, the definition of the periods for trends calculations were broadly in accordance with the implementation of the Euro 4 for new cars (in January 2005) and the Euro 5 (September 2009). The implementation of the Euro classes for HGVs and buses took place in October 2005 (Euro IV), in October 2008 (Euro V) and in January 2013 (Euro VI).

Linear trends over the five-year-periods were calculated using the Theil-Sen method (Theil, 1950; Sen, 1968) available in the R-Openair package (version 1.8–3) (Carslaw and Ropkins, 2011). Briefly, given a set *n x,y* pairs, the slopes between all pairs of points are calculated and the median is given as the most probable slope (trend). This method is robust to outliers and can be used in non-normal and heteroscedastic data series. Confidence intervals at the 95% and the p-values were calculated by bootstrap sampling. A statistically significant trend was assumed when $p < 0.1$ (represented with a '+' symbol), meaning that the trend was not random at a 90% chance; $p < 0.05$, $p < 0.01$ and $p < 0.001$ (marked by '*', '**' and '***', respectively) indicate very highly significant trends; and $p > 0.1$ indicate insignificant trends.

Trends in roadside increments were calculated from monthly means that were first calculated from hourly roadside increments with data capture greater than 75%. A minimum data capture of 75% was imposed to calculate a valid aggregated value (EEA, 2014). Missing monthly data was then linearly interpolated. Time series were de-seasonalized by applying a LOESS smoothing function (Cleveland et al., 1990). Only sites with at least 45 months of available data for each of the five-year-periods (75% data capture) were reported. A sensitivity test in the trend calculations was carried out to evaluate the effect of the data capture threshold and the possible effect of autocorrelation in the time series. Details are given in Supplementary Table 1.

The overall trend for all roadside and kerbside AQMSs in London for each time period was calculated by fitting the linear Random-Effects Model "DerSimonian-Laird estimator" (from the R/metafor package; Viechtbauer, 2010). The Random-Effects (RE) fit assumes that there are two sources of variation in the data set: the within-site estimation variance (variability in the trend calculated for one site as expressed by the confidence intervals) and between sites (variability of trends among the population of sites) (Borenstein et al., 2010). All trends for sites with more than 45 months in each 5-year period of data were included in the calculation of the overall trend regardless of their significance. The graphical representation of the distribution of trends for all roadside sites in London, along with individual confidence intervals and the overall trend was done through "Forest plots". The Forest plot summarizing the trends in ΔNO_X, ΔNO_2 and ΔPM for the population of AQMSs in London and the resulting overall trend for the period 2010–2014 is shown in Fig. 1 as example.

The overall trend was expressed as a percentage by dividing the overall trend by the overall annual mean increment in the first year. This latter was also calculated using the RE fit. Individual variances for the annual mean increment were calculated using the propagation of errors methodology using the uncertainty of the instruments used in the calculation of increments: 4% for NO_X and NO_2 measurements; 6.2% and 17% for PM measurements done by BAM and TEOM-FDMS, respectively; 11.9% for CBLK measurements (Butterfiled et al., 2013) and 1.19 ppmv for CO_2 measurements.

Trends in traffic counts were computed as the slope resulting from the least-square linear model of AADF per year and were expressed as $\Delta vehicles \cdot day^{-1} \cdot year^{-1}$. Traffic trends could not be calculated using the Theil-Sen method because only annual values were available (five data points) insufficient for the Theil-Sen approach. Overall trends for AADF were also calculated by means of the RE model.

2.5. Cluster analysis

Given the large number of monitoring sites and in order to group locations with similar responses in recent trends in air pollutants, the *k*-means algorithm was applied. The cluster analysis was performed on the trends in ΔNO_X, ΔNO_2 and ΔPM_{10} in the period 2010–2014 (variables) for all available monitoring sites (observations). Variables were normalized (mean = 0; variance = 1) before clustering. The selection of the number of clusters was based on the majority rule from a set of 30 indices calculated using the R-NbClust package (Charrad et al., 2014).

3. Results

3.1. Recent trends of air pollutants

There was a change in the sign in trends from 2005–2009 to 2010–2014 for ΔNO_X and ΔNO_2. During the first period ΔNO_X and ΔNO_2 increased with a significant trend (at an average rate of 1% $year^{-1}$ and $11\% \cdot year^{-1}$, respectively), but this reversed in the second period (-1% $year^{-1}$ and $-5\% \cdot year^{-1}$, respectively) (Table 1). Trends in ΔPM_{10} decreased at a significant rate in 2005–2009 ($-4\% \cdot year^{-1}$) but remained stable in 2010–2014. Roadside $PM_{2.5}$ concentrations were only available for the second period and showed a fast and significant decrease: $-28\% \cdot year^{-1}$. Trends in ΔCO_2 and ΔCBLK were also calculated for the three AQMS measuring these metrics in 2010–2014: 0.35 ppm $year^{-1}$ ($2.9\% \cdot year^{-1}$) but not significant; and -0.59 μg m^{-3} $year^{-1}$ ($-11.30\% \cdot year^{-1}$), respectively. Trends in $\Delta PM_{2.5}$ for the three CBLK sites were -0.52 μg m^{-3} $year^{-1}$ ($-14.7\% \cdot year^{-1}$).

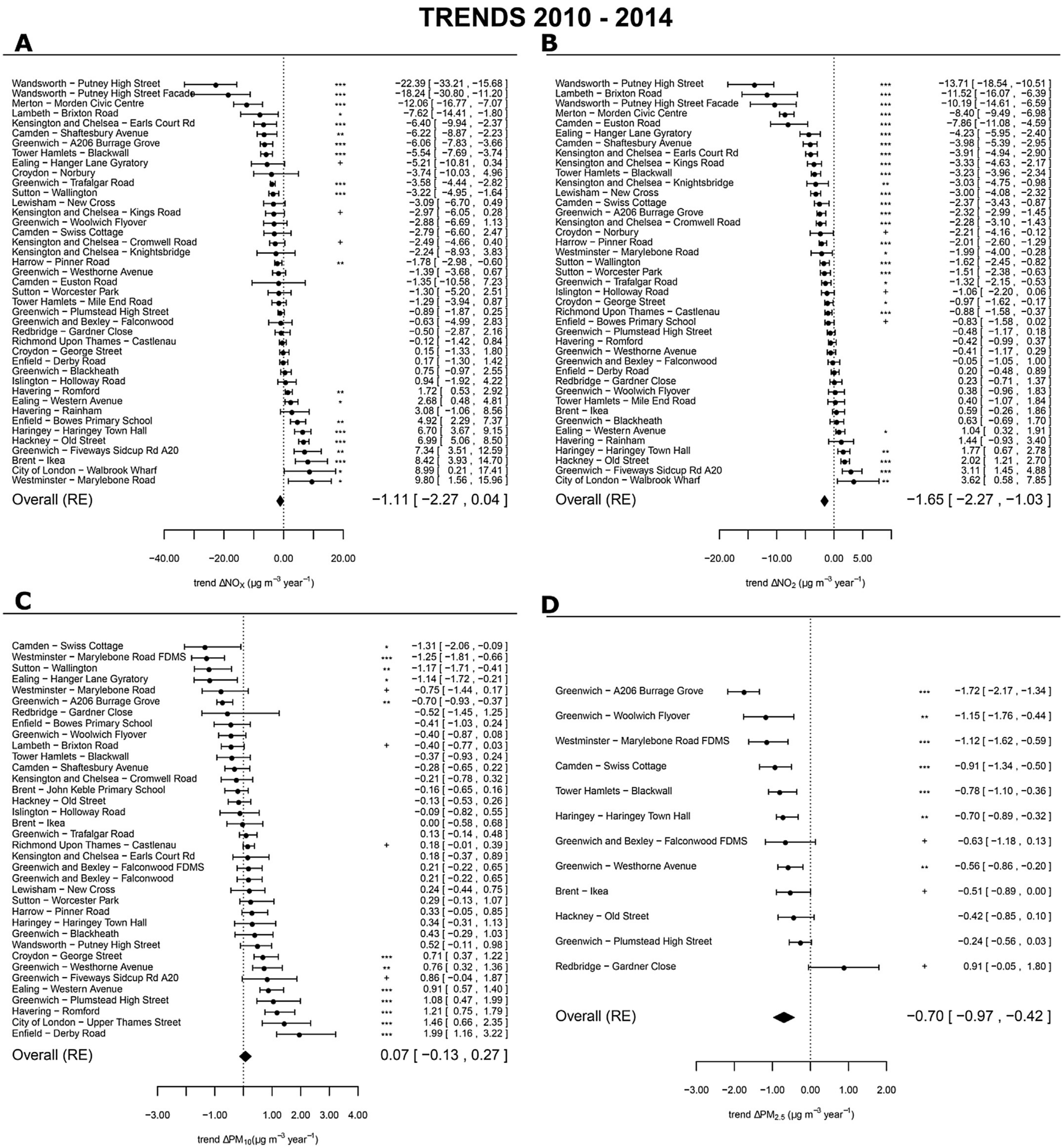

Fig. 1. Forest plots for the trends in roadside increments of NO_X (A), NO_2 (B), PM_{10} (C) and $PM_{2.5}$ (D) for 2010–2014 (in μg m^{-3} $year^{-1}$). *** significant at the 0.001 level; ** significant at the 0.01 level; * significant at the 0.05 level; + significant at the 0.1 level; (blank) not statistically significant. Overall (RE) refers to the overall trend.

Trends in ΔNO_X, ΔNO_2 and ΔPM_{10} at individual roads in London showed more variability during 2010–2014 when compared with 2005–2009 as shown by the wider confidence interval for the overall trends (Table 1). The downward trend for ΔNO_2 was exhibited at more sites (29 of which 25 were statistically significant) than for ΔNO_X (27 of which 14 were statistically significant) (Fig. 1A, B). However, some sites exhibited a positive trend in ΔNO_X (14, 9 of which were statistically significant) and in ΔNO_2 (12, 5 statistically significant). For ΔPM_{10} (Fig. 1C) the majority of sites showed a non-statistically significant trend with confidence intervals including zero (20 sites). However, few sites exhibited a significant negative downward trend (7) and some a positive upward trend (9). Trends in $\Delta PM_{2.5}$ (Fig. 1D) showed a more consistent picture across London's roads with a general statistically

Table 1
Overall absolute and percentage trend calculated by means of the random-effects linear model for the roadside increments (Δ) in NO_X, NO_2, PM_{10}, $PM_{2.5}$, CBLK and CO_2 for the periods 2005–2009 and 2010–2014. Brackets denote 95% confidence intervals.

Pollutant	Overall trend	2005–2009	2010–2014
ΔNO_X	$\mu g\ m^{-3}\ year^{-1}$	0.87 [0.07, 1.68]	−1.11 [−2.27, 0.04]
	% $year^{-1}$	1.02 [0.07, 1.96]	−0.95 [0.04, −1.94]
ΔNO_2	$\mu g\ m^{-3}\ year^{-1}$	1.63 [1.25, 2.01]	−1.65 [−2.27, −1.03]
	% $year^{-1}$	10.56 [8.08, 13.04]	−4.84 [−2.98, −6.69]
ΔPM_{10}	$\mu g\ m^{-3}\ year^{-1}$	−0.19 [−0.34, −0.03]	0.07 [−0.13, 0.27]
	% $year^{-1}$	−3.92 [−0.69, −7.15]	1.11 [−2.06, 4.27]
$\Delta PM_{2.5}$	$\mu g\ m^{-3}\ year^{-1}$	–	−0.70 [−0.97, −0.42]
	% $year^{-1}$	–	−28.34 [−14.65, −42.03]
ΔCO_2	$\mu g\ m^{-3}\ year^{-1}$	–	0.35 [−0.42, 1.11]
	% $year^{-1}$	–	2.93 [−4.00, 9.85]
ΔCBLK	$\mu g\ m^{-3}\ year^{-1}$	–	−0.59 [−0.96, −0.23]
	% $year^{-1}$	–	−11.30 [−3.44, −19.16]

significant downward trend with the single exception of Redbridge – Gardener Close (RB4) which showed an upward trend.

The rate of reduction in ΔNO_2 and ΔNO_X was similar in most AQMSs between 2010 and 2014 (Fig. 2A) with most the sites aligned on the 1:1 line. Sites located in the right bottom quadrant in Fig. 2A experienced a downward trend for ΔNO_2 concentrations whilst ΔNO_X increased. The increased roadside NO offset any gain obtained from the downward trend in roadside NO_2. Alongside those roads in the top right quadrant in Fig. 2A, the increase ΔNO_2 was at a lower rate than the change ΔNO_X indicating that both ΔNO and ΔNO_2 increased.

The comparison between the trends in $\Delta PM_{2.5}$ and in ΔPM_{10} indicated that the majority of sites in inner London experienced a downward trend in both PM fractions at similar rates. With the exception of Greenwich – A206 Burrage Grove (GN0), sites in outer London experienced an increase in ΔPM_{10} while $\Delta PM_{2.5}$ decreased (sites in the right bottom quadrant in Fig. 2B); implying an increase in coarse PM fraction whilst the levels in fine fraction went down. Only Redbridge – Gardner Close (RB4) experienced an increase in $\Delta PM_{2.5}$ while ΔPM_{10} levels decreased.

Trends in ΔCO_2 showed the opposite trend to ΔPM_{10} and $\Delta PM_{2.5}$ (while ΔPM concentrations decreased over time, ΔCO_2 increased; Fig. 2C, D) and did not match those of ΔNO_X and ΔCBLK (Fig. 2E, F). The decrease in ΔCBLK was consistent with the decrease in ΔPM_{10} and $\Delta PM_{2.5}$ (Fig. 2G, H) with trends aligned along the 1:1 line.

3.2. Trends in traffic counts

An overall significant decrease in total vehicles and cars and taxis was observed in 2005–2009 and 2010–2014 on those roads where measurements of both NO_X and PM_{10} were available; a mean rate of −1.0 and $-0.5\%\cdot year^{-1}$ for total vehicles; and −1.3 and $-0.6\%\cdot year^{-1}$ for cars for each time period, respectively (Table 2). HGVs, LGVs and motorcycles decreased in 2005–2009 but only the first was statistically significant. Buses and coaches observed a fast increase in 2005–2009 at $3.2\%\cdot year^{-1}$. Conversely, buses and coaches decreased in 2010–2014, along with motorcycles and LGVs although the later were not statistically significant. Conversely, an overall significant increase in HGVs was observed at a rate of $1.7\%\cdot year^{-1}$ in 2010–2014.

3.3. Cluster analysis

The *k-means* algorithm was used to group sites with similar trends in the 2010–2014 period when the largest variability in responses across the network was observed. A sensitivity test indicated that two sites that had very negative trends in ΔNO_2 (Wandsworth – Putney High Street (WA7) and Lambeth – Brixton Road (LB4)) (Fig. 2A) were unduly affecting the clustering. Furthermore the fleet composition in these sites showed a larger percentage of buses and coaches differentiated them from the other clusters (Supplementary Fig. 5). Both sites were therefore allocated to an *a priori* cluster (cluster #0). Thus 31 monitoring sites were used in the cluster analysis. The monitoring sites were separated into three main clusters containing 15, 7 and 9 sites, respectively (Table 3). The traffic composition in clusters #1 to #3 was very similar (Supplementary Fig. 5) but sites in cluster #2 had the highest traffic counts in the network (a median of 52,102 vehicles·day^{-1} in 2014) while clusters #1 and #3 had median counts of 25,749 and 29,587 vehicles·day^{-1}, respectively (Table 3). There was a general decrease in total traffic between 2010 and 2014 in all clusters although for cluster #0 and cluster #2 the decrease was not statically significant (Fig. 3; Supplementary Table 2).

Downward trends in ΔNO_X were observed on roads in cluster #0 and cluster #3 at similar rates (−3.8 and $-3.3\%\cdot year^{-1}$, respectively); and in both cases accompanied by a significant downward trend in ΔNO_2 ($-9.6\%\cdot year^{-1}$ and $-7.1\%\cdot year^{-1}$, respectively) (Table 3). Cluster #1 also observed a downward trend in ΔNO_X ($-0.7\%\cdot year^{-1}$) and in ΔNO_2 ($-3.4\%\cdot year^{-1}$) but the first was not statistically significant. Generally, the decrease in total traffic was accompanied by a decrease in ΔNO_X and ΔNO_2 on these roads (Fig. 3a). In sharp contrast we found that sites in cluster #2 observed an overall positive trend in ΔNO_x ($5.6\%\cdot year^{-1}$, statistically significant) and in ΔNO_2 ($2.7\%\cdot year^{-1}$, not statistically significant) (Table 3). The increase in ΔNO_X and ΔNO_2 observed on these roads (Fig. 4a,b; Fig. 5a,b) might be related to upward trends in buses and coaches and HGVs. The correlation (R^2) between trends in ΔNO_X and that in buses and coaches (all sites) was 0.29 ($p < 0.01$). Hackney – Old Street (HK6) was an exception and the increase in ΔNO_X and ΔNO_2 occurred with an increase in cars and taxis (Supplementary Figs. 4b and 5b) and motorcycles (Supplementary Figs. 4c and 5c).

Only two clusters showed significant trends in ΔPM_{10} in 2010–2014: cluster #1, with a positive trend at $11.9\%\cdot year^{-1}$; and cluster #3, with a downward trend of $-6.3\%\cdot year^{-1}$. In both clusters, the trends in $\Delta PM_{2.5}$ showed a negative trend at −20.5 and $-28.6\%\cdot year^{-1}$, respectively. Sites in cluster #3 observed a general decrease in both fractions as traffic flows reduced (Fig. 3c and d). The positive trends in ΔPM_{10} observed on roads in cluster #1 were mostly associated with increased HGVs (Fig. 5c). Sites in cluster #2 also observed a significant downward trend in $\Delta PM_{2.5}$ ($-32.5\%\cdot year^{-1}$) despite trends in ΔPM_{10} not showing a clear tendency. Some roads in cluster #2 with increased HGVs observed a negative trend in ΔPM_{10} (Westminster - Marylebone Road (MY1, MY7) and Tower Hamlets – Blackwall (TH4)) in contrast to that observed in roads in cluster #1.

4. Discussion

There was an overall decrease in total traffic between 2005 and 2014, mainly explained by the reduction in cars and taxis. This concurs with the “peak car” phenomena observed in many cities worldwide (Puentes and Tomer, 2008; Metz, 2013). In London, car use declined despite an increase in population in recent years (Metz, 2015; Focas, 2016). This might be explained by constraints on road capacity for new cars and also by the re-allocation of road space to buses, cycle lanes and pedestrians (TfL, 2014b). However, despite the reduced traffic, levels of roadside pollutants did not decrease accordingly.

The increase in NO_X and in NO_2 roadside concentrations during 2005–2009 is in sharp contrast to the decreasing trends in traffic and also to the predicted impacts of tighter Euroclass emissions

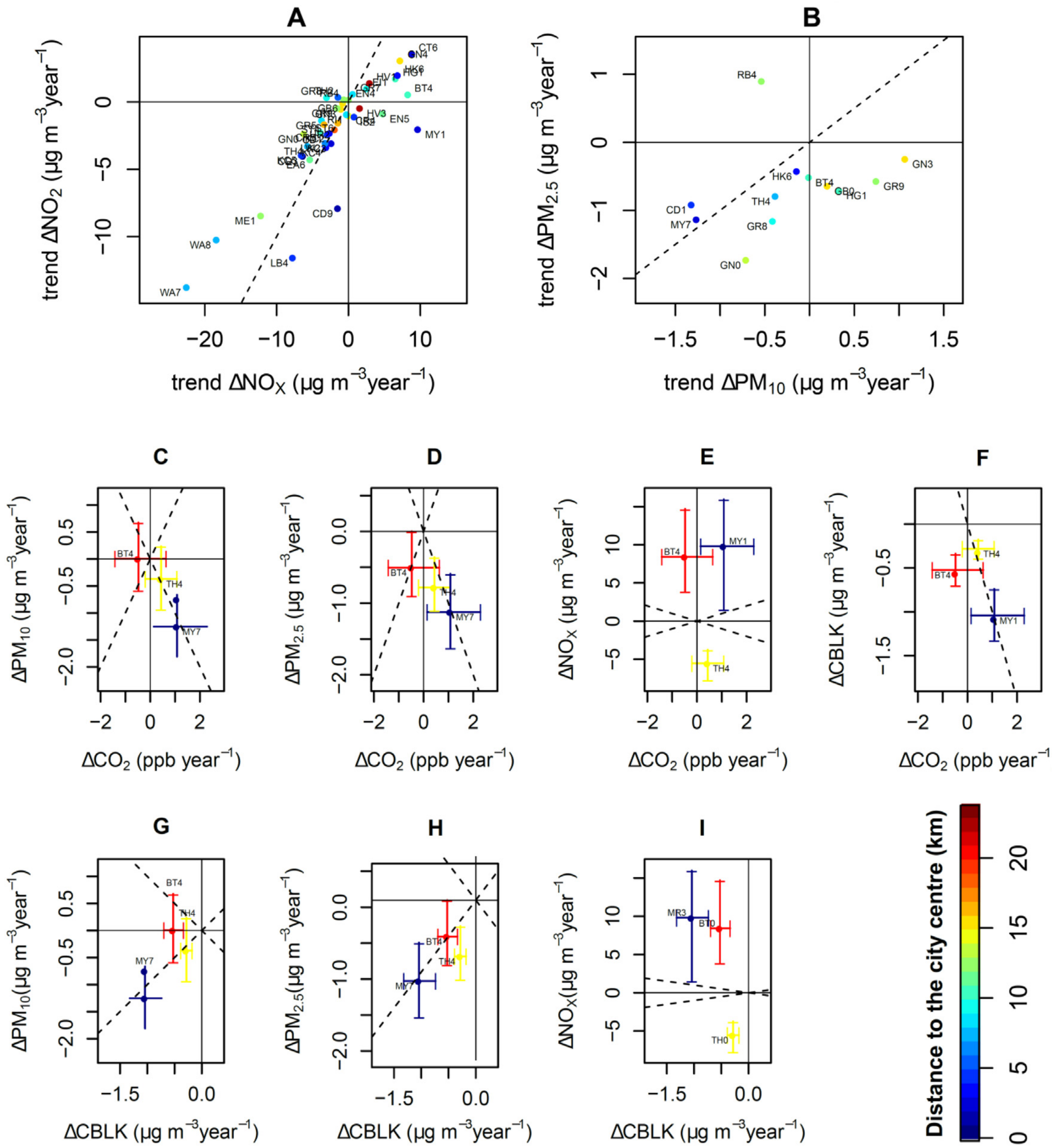

Fig. 2. Trends in ΔNO_2 vs trends in ΔNO_X (A); trends in $\Delta PM_{2.5}$ vs trends in ΔPM_{10} (B); trends in ΔCO_2 increments (C–E) and trends in roadside black carbon increments (G–I) against trends in the regulated pollutants for the period between 2010 and 2014; and comparison of trends in roadside black carbon against trends in roadside CO_2 (F). Dashed lines indicates the 1:1 and the −1:1 relationships.

Table 2
Overall absolute and percentage trend calculated by means of the random-effects linear model for the vehicles counts for 2010–2014. Brackets denote 95% confidence interval.

Vehicle category	Overall trend	2005–2009	2010–2014
Total vehicles	Δvehicles day^{-1} year^{-1}	−402.64 [−557.48, −247.81]	−196.56 [−286.36, −106.77]
	% year^{-1}	−1.05	−0.50
Cars & taxis	Δvehicles day^{-1} year^{-1}	−371.49 [−500.33, −242.66]	−167.41 [−247.14, −87.68]
	% year^{-1}	−1.28	−0.56
Buses & coaches	Δvehicles day^{-1} year^{-1}	36.89 [26.22, 47.56]	−9.14 [−21.11, 2.83]
	% year^{-1}	3.21	−0.71
Motorcycles	Δvehicles day^{-1} year^{-1}	−3.04 [−8.86, 2.79]	−5.42 [−11.28, 0.44]
	%·year^{-1}	−0.26	−0.43
HGVs	Δvehicles day^{-1} year^{-1}	−11.47 [−21.73, −1.20]	29.48 [14.17, 44.78]
	% year^{-1}	−0.63	1.69
LGVs	Δvehicles day^{-1} year^{-1}	−2.33 [−24.17, 19.50]	−19.25 [−47.30, 8.79]
	% year^{-1}	−0.05	−0.38

Table 3
Overall trend in ΔNO_X, ΔNO_2 and ΔPM_{10} for each cluster.

Cluster #	0	1	2	3
N sites	2	15	7	9
Trend ΔNO_X ($\mu g\ m^{-3}\ year^{-1}$)	−14.70 [−29.16, −0.23]	−0.41 [−1.45, 0.64]	6.66 [5.51, 7.81]	−4.52 [−5.62, −3.43]
Trend ΔNO_X (% $year^{-1}$)	−3.81 [0.08, −7.71]	−0.70 [1.08, −2.47]	5.62 [3.47, 7.77]	−3.33 [−2.22, −4.43]
Trend ΔNO_2 ($\mu g\ m^{-3}\ year^{-1}$)	−12.82 [−15.85, −9.79]	−0.48 [−0.92, −0.05]	0.81 [−0.52, 2.13]	−2.89 [−3.45, −2.33]
Trend ΔNO_2 (% $year^{-1}$)	−9.59 [−7.28, −11.90]	−3.41 [−0.22, −6.61]	2.70 [−1.84, 7.25]	−7.06 [−4.88, −9.25]
Trend ΔPM_{10} (% $year^{-1}$)	0.05 [−0.86, 0.95]	0.46 [0.21, 0.70]	−0.23 [−0.69, 0.23]	−0.48 [−0.80, −0.16]
Trend ΔPM_{10} (% $year^{-1}$)	0.46 [−7.84, 8.75]	11.95 [5.28, 18.61]	−2.51 [2.59, −7.60]	−6.27 [−1.87, −10.68]
Trend $\Delta PM_{2.5}$ (% $year^{-1}$)	–	−0.40 [−0.84, 0.03]	−0.67 [−0.93, −0.42]	−1.13 [−1.70, −0.56]
Trend $\Delta PM_{2.5}$ (% $year^{-1}$)	–	−20.83 [3.19, −44.85]	−32.52 [−10.48, −54.56]	−28.61 [−10.57, −46.65]
Distance to city centre (km) (mean ± sd)	6.51 ± 2.16	12.98 ± 4.48	8.16 ± 5.23	8.09 ± 5.16
Median AADF in 2014 (# vehicles·day^{-1})	21,036	25,749	52,102	29,587
List of AQMSs	WA7, LB4	CR4, EI1, EN4, GB6, GN3, GR5, GR7, GR8, GR9, HR2, HV3, IS2, RB4, RI1, ST6	BT4, EN5, GN4, HG1, HK6, MY1, MY7	CD1, CD3, EA6, GN0, KC2, KC5, LW2, ST4, TH4

factors. But it concurs with a growing body of evidence that suggests that real-world emissions from diesel vehicles did not align with improved performance found in approval tests. Diesel emissions make important contributions to NO_X and primary NO_2 emissions in urban areas (Sundvor et al., 2012 and references within). In 2005–2009 the number of diesel vehicles increased by 33% for the whole of the UK (DfT, 2015). The Catalytic Diesel Particle Filters (CDPF) in introduced in Euro 4 cars to reduce PM emissions from 2005 onwards were proved to emit higher NO_2/NO_X ratios than those not fitted with CDPF leading to an increase in roadside NO_2 concentrations (Carslaw et al., 2006). In London, part of the diesel emissions are attributed to buses, especially in some central routes where diesel powered busses are estimated to contribute 33% of the total road transport NO_2 emissions (GLA, 2013; Carslaw et al., 2015). During 2005–2009, buses and coaches increased at a rate of 3%·$year^{-1}$, contributing to the increase in roadside NO_X and NO_2 concentrations.

In 2005–2009 an overall decrease in PM_{10} concentration was observed on the majority of roads across London. One of the possible explanations is the efficacy of new CDPF; another is the general decrease in HGVs in this period reducing non-exhaust traffic emissions from resuspension, brake and tyre-wear.

Overall roadside NO_X and NO_2 decreased in 2010–2014, along with $PM_{2.5}$. The decrease in these pollutants across London's roads might be partly explained by the general decrease in total vehicles since 2010, at about 0.5%·$year^{-1}$. However, some roads observed an increase in their roadside NO_X and NO_2 concentrations despite the decreased traffic flow (Fig. 3a, b). The dieselization of the vehicle fleet continued during this period, increasing by 25% (DfT, 2015). Despite the introduction of Euro 5 in cars in September 2009 which set lower NO_X emission limits (28% and 25% lower for new diesel and petrol cars, respectively, when compared with Euro 4) remote sensing measurements of exhaust plumes from diesel Euro 5 cars in London indicated that under real-driving conditions emissions were similar to Euro 4 (Carslaw et al., 2016). The same pattern was observed for diesel LGVs. Interestingly, remote testing campaigns also indicated that emissions of primary NO_2 from diesel Euro 4 and Euro 5 cars reduce they age (Carslaw et al., 2016). This might explain the reduction observed across London rather than lower emissions from the newest cars.

The introduction of the Low Emissions Zone in 2008, which banned the most polluting diesel HGVs and LGVs, might have also contributed to the general reduction in NO_X and NO_2. In October 2008 Euro V were introduced for HGVs which tightened NO_X emissions by 42% compared to Euro IV. NO_2/NO_X ratios under real-driving conditions were similar for both Euro IV and V but both were clearly better than Euro III (Carslaw et al., 2016). Some sites observed an increase in their HGVs while NO_2, NO_X (and also $PM_{2.5}$) concentrations went down (Fig. 5a,b) indicating the improvement in emission rates in this type of vehicles: Tower Hamlets – Blackwall (TH4), Greenwich – Trafalgar Square (GR5) and Greenwhich – Westhorne Avenue (GR9), among others.

Despite the general downward trend in roadside NO_X and NO_2 in 2010–2014 there was large heterogeneity across London's roads. The comparison of trends for the different pollutants measured at the same AQMS offers an insight into the changes in sources and the success of abatement policies. The comparison of trends in ΔNO_2 against ΔNO_X indicated that the reduction in ΔNO_X was mostly explained by the reduction in ΔNO_2 for the majority of roads with downward trends suggesting that abatement policies mostly tackled NO_2. Given that NO to NO_2 conversion by ozone (O_3) is limited by O_3 availability close to roads (Carslaw and Beevers, 2005), this decrease in roadside NO_2 suggests a decrease in primary NO_2 emissions.

Those roads with increased NO_X and NO_2 also observed increase in buses and coaches (Fig. 4a) and to a lesser extent to HGVs (Fig. 5a). Despite the clear social and environmental benefit of increasing the capacity of public transport, this study shows that an increase in buses and coaches was associated with an increase in ΔNO_X and ΔCO_2; and a lesser extent in ΔNO_2 and ΔPM_{10} (Fig. 4). Improving the emissions standards of buses and coaches is therefore crucial. Retrofitting Euro III buses with low-NO_2 Selective Continuous Regeneration Trap (SCRT) systems have been shown to reduce primary emissions of NO_2 by 61% and NO_X by 45% under real-driving conditions in London compared to buses only fitted with CRT (Carslaw et al., 2015). Retrofitting buses along Putney High Street led to a decrease in local NO_X and NO_2, with the concentration of NO_X decreasing to a greater extent than that of NO_2 (Barratt and Carslaw, 2014) consistent with the patterns seen in our analysis (WA7, WA8). This technology might be beneficial on other roads with positive trends in ΔNO_X associated with an increase in buses and coaches such as Westminster-Marylebone Road (MY1), Haringey – Town Hall (HG1) and Ealing – Westbourne Avenue (EI1).

Less traffic should mean less exhaust traffic emissions (as observed in roads in cluster #3). However, some sites observed a decrease in some air pollutants while traffic increased indicating an improvement in the emission standards. This was the case for $PM_{2.5}$ which benefitted from control on exhaust emissions in different vehicle categories (Supplementary Fig. 10); and for CBLK that decreased despite an increase in the total number of vehicles and buses and coaches (Figs. 3f and 4f, respectively). However, the clear benefit of controlling exhaust emissions can be offset by increased non-exhaust emissions with more traffic in the roads, especially heavy vehicles. This is the case for those roads with increased PM_{10} and HGVs (roads in cluster#1, Supplementary Table 5). Despite the

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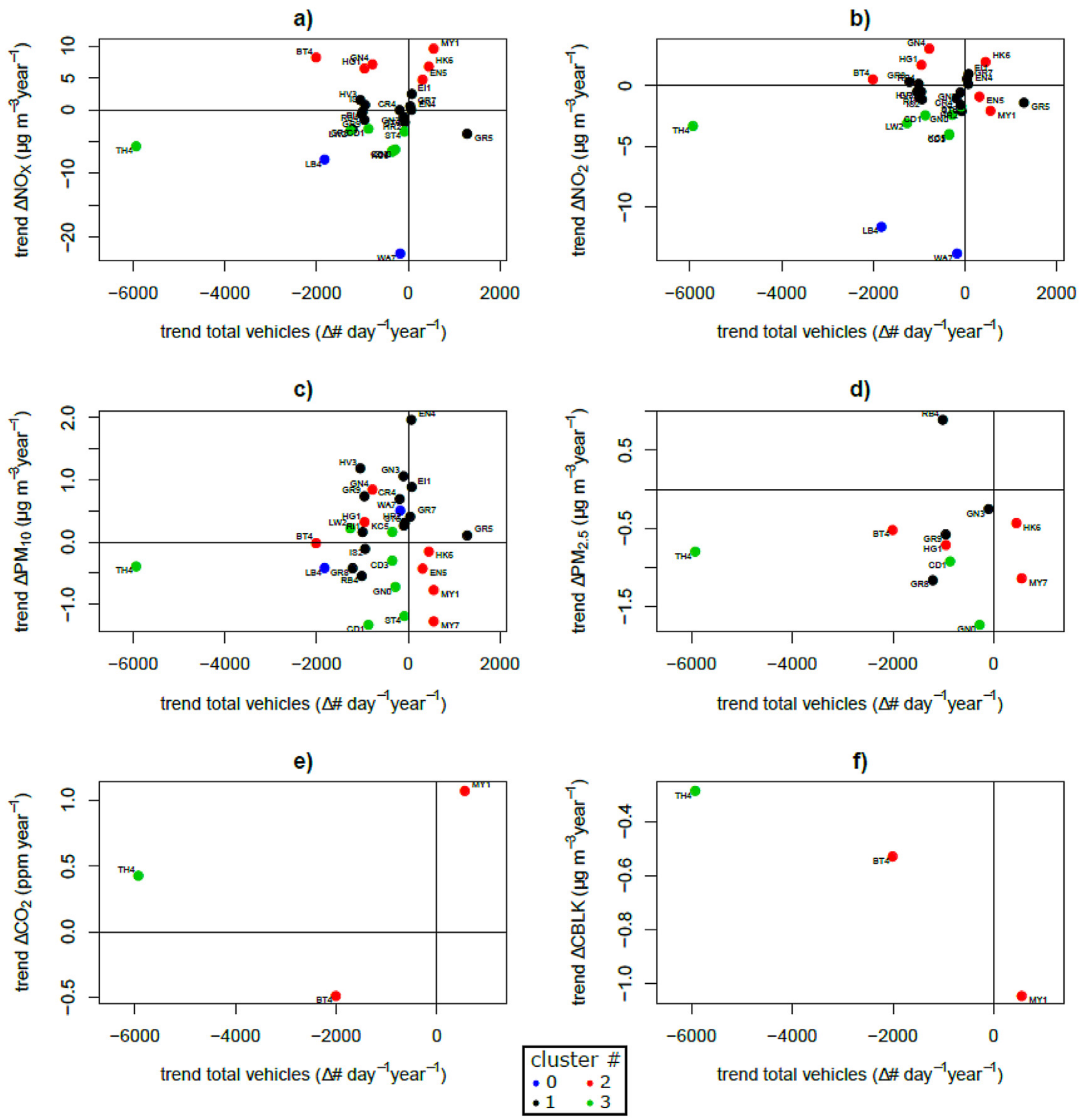

Fig. 3. Trends in air pollutants versus trends in total vehicles for 2010 to 2014. Colour indicates site clusters. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

improvement in the emissions of fine particulate matter from HGVs in London (Fig. 5d), an increase in these vehicles resulted in an increase in the coarse PM fraction which is associated with non-exhaust traffic emissions such as resuspension, tyre-wear and brake-wear. Enhancement of the coarse PM fraction was previously observed in central roads in London with an increase in the number of buses following the introduction of the Congestion Charge in 2003 (Carslaw et al., 2006). With no emission control strategies taken by the EU member states, non-exhaust traffic emissions have become a very important source of PM, (van der Gon et al., 2013; Amato et al., 2014a), and these can contribute to a large proportion of PM exceedances in urban areas (Harrison et al., 2014). Road dust is made of harmful components such as heavy metals (Amato et al., 2009), sulphides, carbonaceous aerosols and Polycyclic Aromatic Hydrocarbons (Pengchai et al., 2004; Majumdar et al., 2012). Regulation of non-exhaust emissions of PM is therefore crucial. Some actions have attempted to reduce non-exhaust emissions such as the application of dust binders such as Calcium Magnesium Acetate (CMA). The effectiveness of dust binding has been shown in Sweden (Norman and Johansson, 2006) and Austria (www.life-cma.at) with daily PM_{10} decreased up to 35%. However, other studies in Germany (Reuter, 2010), United Kingdom (Barratt et al., 2012) and Spain (Amato et al., 2014b) could not detect a significant PM_{10} decrease on typical urban roads. PM_{10} did not increase on all roads with more HGVs: Westminster - Marylebone Road (MY1, MY7) and Tower Hamlets – Blackwall (TH4) (Fig. 5c). These roads are usually more congested than those in outer London (such as sites in cluster #1) suggesting that other factors such as traffic speed and type of flow (stop-go vs freeway driving) might also influence the non-exhaust emission rates.

The trend in ΔCO_2 showed an unexpected behaviour. The London Atmospheric Emissions Inventory (LAEI) predicted ~ 1% annual decrease in road transport CO_2 emissions between 2010 and 2014. This was not borne out by roadside measurements which showed an overall increase of 2.9%$\cdot$year^{-1}. ΔCO_2 was expected to decrease with reduced traffic counts, improving fuel fleet efficiency and due to the increase of alternatively-fuelled vehicles. Positive trends in ΔCO_2 in 2010–2014 were observed on those roads with more buses

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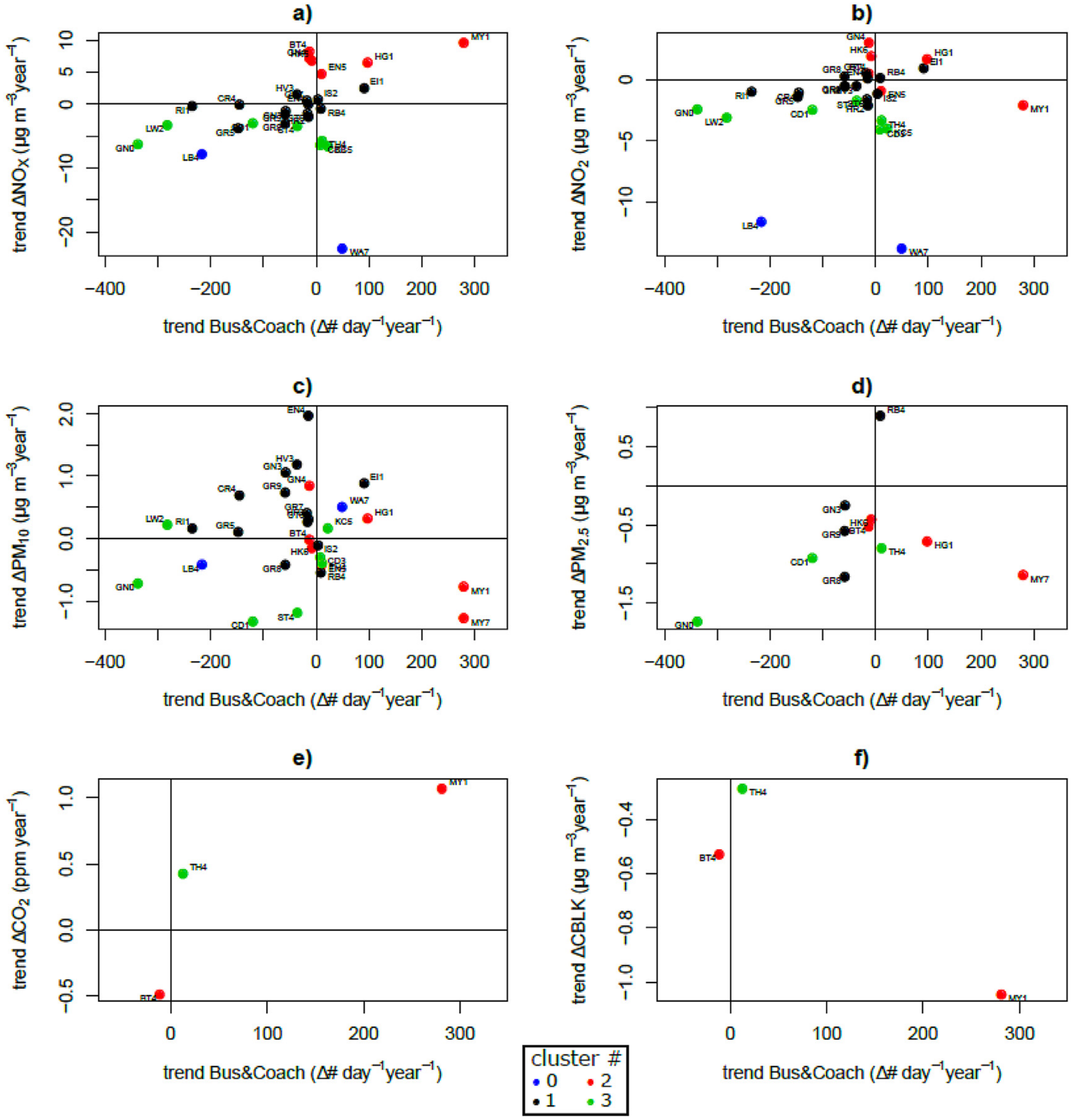

Fig. 4. Trends in air pollutants versus trends in buses and coaches for 2010 to 2014. Colour indicates site clusters. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

and HGVs (Figs. 4e and 5f). Some authors have highlighted that the use of particulate filters on the vehicles can led to increased fuel consumption and hence more CO_2 emissions (Monks et al., 2009). However, it is expected that CO_2 emissions from these vehicles will reduce in the coming years in line with the European Heavy-Duty Vehicles strategy, adopted in May 2014. It should be noted that our methodology to calculate ΔCO_2 did not account for non-anthropogenic emissions (e.g. overnight vegetation respiration) that might affect background locations (Priestman et al., in prep).

5. Conclusions

Here we used a population of monitoring sites to examine trends of air pollutants between 2005–2009 and 2010–2014 in London taking into account both individual and population-wide variability. Policies that aimed to reduce ambient air pollution levels by regulating traffic behaviour and emissions in London had a clear impact from 2010 onwards. The majority of roadside and kerbside sites in London had a significant downward trend in their roadside increment for NO_2 (8–13%$\cdot$year^{-1}) and $PM_{2.5}$ (15–45%$\cdot$year^{-1}). This contrasted with the trends for the preceding five year period (2005–2009) when a wider upward tendency was observed in ΔNO_2 concentrations. No data was available to compare trends in $\Delta PM_{2.5}$ between the two periods.

The overall downward trend in NO_X and NO_2 in 2010–2014 was likely due to a variety of factors including a general decrease in the traffic flow; a reduction of primary NO_2 emissions from aging diesel passenger cars; and the introduction of the Low Emissions Zone with cleaner HGVs.

Roadside $PM_{2.5}$ and CBLK decreased at similar rates on roads with collocated measurements. These decreases were attributed to the effectiveness of diesel particle filters. However, the number of black carbon measurement sites was small and an extended number of sites will be needed to assess this fully. Using a filter based technique Davy (2014) found increases in black carbon alongside two central London roads between 2011 and 2013. Given the heterogeneity in the behaviour of other pollutants we should be cautious about concluding that these technologies are being

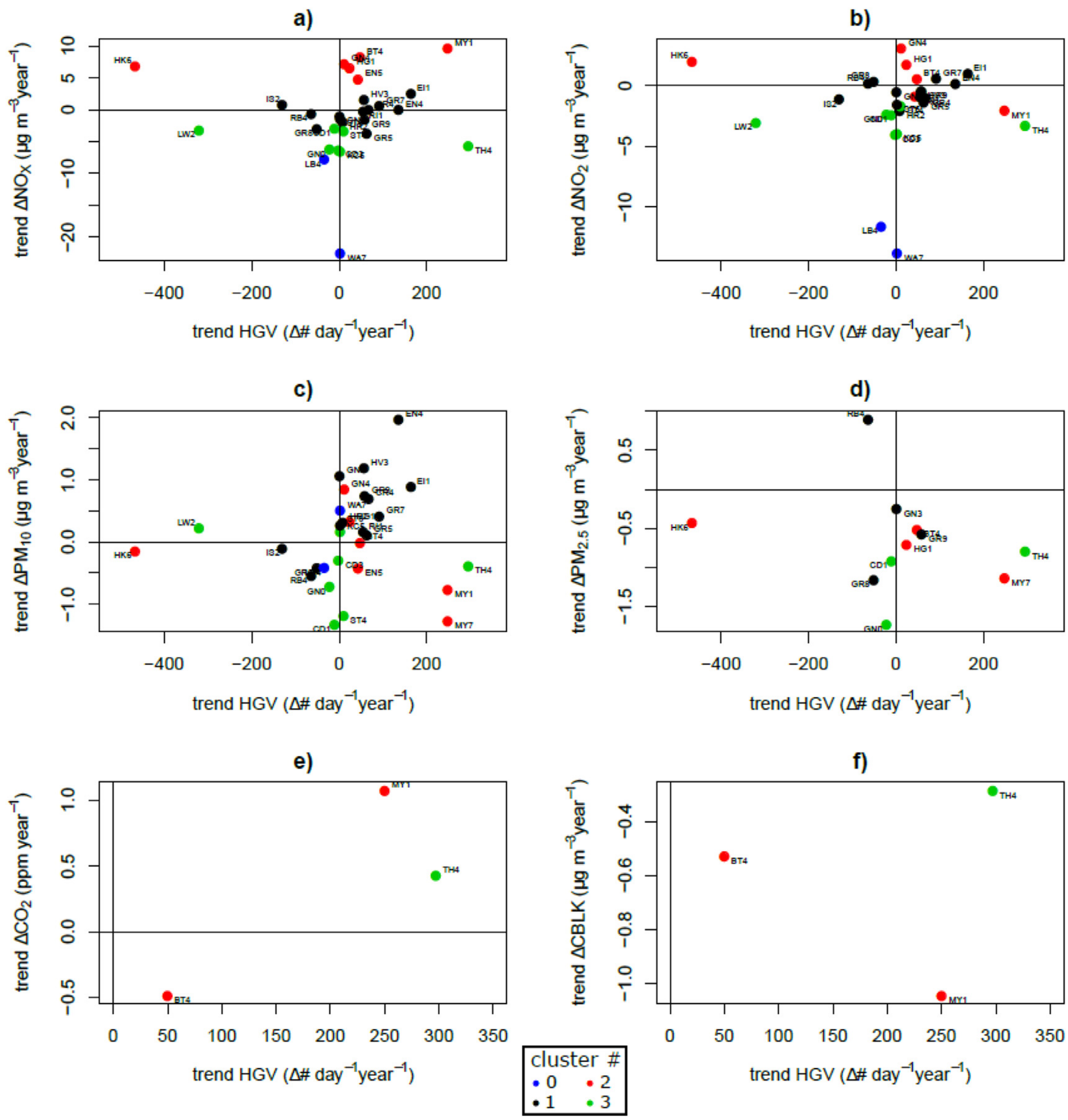

Fig. 5. Trends in air pollutants versus trends in HGVs for 2010 to 2014. Colour indicates site clusters. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

effective everywhere.

Separating inner and outer London roads did not group AQMSs according to the observed trends. Therefore the *k*-means cluster analysis was used to group those roadside with a similar policy response. Three relevant clusters of roadside AQMS resulted from the analysis. Two clusters grouped busy roads with median AADF ~25–30·10^3 vehicles·day^{-1} with downward trends in both ΔNO_2 and $\Delta PM_{2.5}$ but with different behaviour in PM_{10}. While one cluster observed a decreased in PM_{10}, the other one experienced a stabilization/increase associated with more coarse PM. The third cluster grouped very busy roads (median AADF > 50·10^3 vehicles·day^{-1}) characterized by an increase in NO_X. These roads had an increase in the number of buses and coaches and at a lesser extent, in HGVs.

Heavy vehicles were a clearly an important factor in urban air pollution. Whilst increased buses might be desirable for many social and environmental reasons it is clear that this has to be in conjunction with investment in cleaner emissions technologies such as the successful installation of low NO_2 SCRT on some of London's buses. Greater management of HGVs is also needed to ensure that increased numbers do not offset benefits from emissions abatement and to control the increase in coarse PM. A greater investigation of sources of PM coarse, the factors which control emission rates and options for managing them is needed.

Attaining the NO_2 EU Limit Value still remains a challenge along most major roads on London. Despite the general downward trend in roadside increments in the period 2010–2014 in London, around three quarters of road and kerbside AQMSs exceeded the NO_2 EU Limit Value in 2015, with seven AQMSs measuring concentrations that were more than twice the limit. Modelling results indicate that London will have difficulties complying with current legislation before 2030 (Kiesewetter et al., 2014). Some of roads with the greatest reduction in NO_2 (e.g. Wandsworth Putney High Street) were due to one off interventions (retrofitting Euro III buses with low NO_2 SCRT) and the reduction rate between 2010 and 2014 might not be a predictor of future trends.

This study showed that there was considerable heterogeneity in the outcome of policy interventions to control air pollution in London's roads and this is likely to be the case in other cities. This highlights the need for detailed measurement and to feedback into the policy making process. In some locations emissions abatement policies were offset by changes in traffic flow. Abatement technologies for tail-pipe emissions might be beneficial to control fine particulate and black carbon emissions from diesel; Euro 6 standards for diesel cars are also expected to lead to decreased NO_X and primary NO_2 emissions under real driving conditions. However, increasing traffic flows especially of those from heavy vehicles will enhance non-exhaust emissions. This suggests that stronger policy packages are therefore needed to control both exhaust and non-exhaust traffic emissions to ensure that all areas, and therefore all communities, benefit from improved air pollution (Fecht et al., 2015). Given that the pollution trends in London are partially the result of European-wide policies, our study suggests that there is an urgent need for detailed analysis across other European cities and regions to ensure that these policies are working well and everywhere.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.envpol.2016.07.026.

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